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GAS SEPARATION FOR MOLTEN CARBONATE FUEL CELL

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FIELD OF THE INVENTION

The present invention relates to a fuel cell-based electrical generation system which employs pressure swing adsorption for enhancing the efficiency of a molten carbonate fuel cell.

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BACKGROUND OF THE INVENTION

Fuel cells provide an environmentally friendly source of electrical current. One form of fuel cell used for generating electrical power, particularly envisaged for larger scale stationary power generation, includes an anode channel for receiving a flow of hydrogen gas, a cathode channel for receiving a flow of oxygen gas, and a porous matrix containing a molten carbonate electrolyte which separates the anode channel from the cathode channel. Oxygen and carbon dioxide in the cathode channel react to form carbonate ions which cross the electrolyte to react with hydrogen in the anode channel to generate a flow of electrons. As the hydrogen is consumed, carbon monoxide is shifted by steam to generate additional hydrogen. Carbon dioxide and water vapour are produced in the anode channel by oxidation of fuel components, and by reduction of carbonate ions from the electrolyte. Typical operating temperature of molten carbonate fuel cells is about 600° to 650° C.

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Except in the rare instance that hydrogen (e.g. recovered from refinery or chemical process offgases, or else generated from renewable energy by electrolysis of water) is directly available as fuel, hydrogen must be generated from fossil fuels by an appropriate fuel processing system. For stationary power generation, it is preferred to generate hydrogen from natural gas by steam reforming or partial oxidation to produce "syngas" comprising a mixture of hydrogen, carbon monoxide, carbon dioxide, steam and some unreacted methane. As hydrogen is consumed in the fuel cell anode channel, much of the carbon monoxide reacts with

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steam by water gas shift to generate more hydrogen and more carbon dioxide. Other carbonaceous feedstocks (e.g. heavier hydrocarbons, coal, or biomass) may also be reacted with oxygen and steam to generate syngas by partial oxidation or autothermal reforming. The fuel cell may also be operated on hydrogen or syngas that has been.

A great advantage of molten carbonate fuel cell (MCFC) systems is that their high operating temperature facilitates close thermal integration between the fuel cell and the fuel processing system. The high temperature also allows the elimination of noble metal catalysts required by lower temperature fuel cells.

Prior art molten carbonate fuel cells also have serious limitations associated with their high temperature operation, and with their inherent need to supply carbon dioxide to the cathode while removing it from the anode.

The lower heat of combustion of a fuel usefully defines the energy (enthalpy change of the reaction) that may be generated by oxidizing that fuel. The electrochemical energy that can be generated by an ideal fuel cell is however the free energy change of the reaction, which is smaller than the enthalpy change. The difference between the enthalpy change and the free energy change is the product of the entropy change of the reaction multiplied by the absolute temperature. This difference widens at higher temperatures, so higher temperature fuel cells inherently convert a lower fraction of the fuel energy to electrical power at high efficiency because a larger fraction of the fuel energy is available only as heat which must be converted by a thermodynamic bottoming cycle (e.g. steam or gas turbine plant) at lower efficiency.

Accumulation of reaction products (carbon dioxide and steam) on the fuel cell anode opposes the electrochemical reaction, so that the free energy is reduced. Higher partial pressure of oxygen and carbon dioxide over the cathode, and higher partial pressure of hydrogen over the anode, drive the reaction forward so that the free energy is increased. Unfortunately, the reaction depletes the oxygen and

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carbon dioxide in the cathode channel and depletes hydrogen in the anode channel while rapidly increasing the back pressure of carbon dioxide in the anode channel. Hence the free energy change is reduced, directly reducing the cell voltage of the fuel stack. This degrades the electrical efficiency of the system, while increasing the heat that must be converted at already lower efficiency by the thermal bottoming cycle.

The free energy change is simply the product of the electromotive force ("E") of the cell and the charge transferred per mole by the reaction ("2F"), where the factor of two reflects the valency of the carbonate ion. The following Nernst relation for a MCFC expresses the above described sensitivity of the electromotive force to the partial pressures of the electrochemical reactants in the anode and cathode channels, where the standard electromotive force ("E_o") is referred to all components at standard conditions and with water as vapour.

$$E = E_o - \frac{RT}{2F} \ln \left[\frac{P_{H_2O(anode)} \cdot P_{CO_2(anode)}}{P_{H_2(anode)} \cdot P_{O_2(cathode)}^{0.5} \cdot P_{CO_2(cathode)}} \right]$$

Prior art MCFC systems do not provide any satisfactory solution for this problem which gravely compromises attainable overall efficiency. The challenge is to devise a method for sustaining high hydrogen concentration over the anode and high oxygen concentration over the cathode, while efficiently transferring hot carbon dioxide from the anode to the cathode. Despite repeated attempts to devise an effective carbon dioxide transfer technology that would be compatible with MCFC operating conditions, no such attempt has been adequately successful.

The accepted method for supplying carbon dioxide to the cathode has been to burn a fraction of the anode gas (including unreacted hydrogen and other fuel components) to generate relatively dilute carbon dioxide mixed with steam and nitrogen to be mixed with additional air providing oxygen to the cathode. This approach has serious limitations. Even more of the original fuel value is unavailable for relatively efficient electrochemical power generation, in view of

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additional combustion whose heat can only be absorbed usefully by the thermal bottoming cycle. Also, the oxygen/nitrogen ratio of the cathode gas is even more dilute than ambient air, further reducing cell voltage and hence transferring more power generation load less efficiently onto the thermal bottoming plant.

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The present invention provides an advanced MCFC system incorporating a high temperature pressure swing adsorption (PSA) and integrated gas turbine system to enrich hydrogen over the anode while rapidly separating carbon dioxide to the cathode. Prior art PSA technology is not capable of operation at such high temperature. Also, adsorbents which can separate carbon dioxide in the presence of steam must be provided.

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PSA systems and vacuum pressure swing adsorption systems (VPSA) separate gas fractions from a gas mixture by coordinating pressure cycling and flow reversals over an adsorber or adsorbent bed which preferentially adsorbs a more readily adsorbed gas component relative to a less readily adsorbed gas component of the mixture. The total pressure of the gas mixture in the adsorber is elevated while the gas mixture is flowing through the adsorber from a first end to a second end thereof, and is reduced while the gas mixture is flowing through the adsorbent from the second end back to the first end. As the PSA cycle is repeated, the less readily adsorbed component is concentrated adjacent the second end of the adsorber, while the more readily adsorbed component is concentrated adjacent the first end of the adsorber. As a result, a "light" product (a gas fraction depleted in the more readily adsorbed component and enriched in the less readily adsorbed component) is delivered from the second end of the adsorber, and a "heavy" product (a gas fraction enriched in the more strongly adsorbed component) is exhausted from the first end of the adsorber.

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However, the conventional system for implementing pressure swing adsorption or vacuum pressure swing adsorption uses two or more stationary adsorbers in parallel, with multiple two-way directional valves at each end of each adsorber to connect the adsorbers in alternating sequence to pressure sources and

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sinks. This system is often cumbersome and expensive to implement due to the large size of the adsorbers and the complexity of the valving required. The valves would not be capable of operation at MCFC working temperatures. Further, the conventional PSA system makes inefficient use of applied energy because of irreversible gas expansion steps as adsorbers are cyclically pressurized and depressurized within the PSA process. Conventional PSA systems are bulky and heavy because of their low cycle frequency and consequent large adsorbent inventory.

A further need addressed by the invention is for mitigation of global warming driven by cumulative emissions of carbon dioxide from fossil fueled power generation. The present invention improves overall efficiency to reduce the proportionate amount of carbon dioxide formed, while enabling delivery as and when desired of that carbon dioxide in highly concentrated form for most convenient sequestration from the atmosphere, e.g. by underground disposal in depleted natural gas reservoirs.

SUMMARY OF THE INVENTION

According to the invention, there is provided a molten carbonate fuel cell based electrical generation system which addresses the deficiencies of the prior art, particularly as to transfer of carbon dioxide from the anode to the cathode while enhancing electrical power output.

The electrical current generating system comprises a molten carbonate fuel cell, an oxygen gas delivery system, and a hydrogen gas delivery system. The fuel cell includes an anode channel having an anode gas inlet for receiving a supply of hydrogen gas, a cathode channel having a cathode gas inlet and a cathode gas outlet, and an electrolyte in communication with the anode and cathode channel for facilitating ion transport between the anode and cathode channel.

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However, for superior performance it incorporates a high temperature PSA system, including a rotary module having a stator and a rotor rotatable relative to the stator, for enriching hydrogen to the anode channel and extracting carbon dioxide therefrom. In some embodiments, the inventive system also includes a lower temperature PSA or VPSA system for enriching oxygen from air for supply to the cathode channel and/or to the fuel processing system. The high temperature PSA unit for enriching hydrogen and separating carbon dioxide will be referred to as the first PSA unit, while a second PSA or VPSA unit may be provided for oxygen enrichment.

The rotor of a PSA unit for the invention includes a number of flow paths for receiving adsorbent material therein for preferentially adsorbing a first gas component in response to increasing pressure in the flow paths relative to a second gas component. The pressure swing adsorption system also may include compression machinery coupled to the rotary module for facilitating gas flow through the flow paths for separating the first gas component from the second gas component. The stator includes a first stator valve surface, a second stator valve surface, and plurality of function compartments opening into the stator valve surfaces. The function compartments include a gas feed compartment, a light reflux exit compartment and a light reflux return compartment.

Operating temperature of the adsorbers in the first PSA unit of the invention may be similar to that of the MCFC, or may be somewhat lower down to about 450° C as may be facilitated by recuperative heat exchange. This PSA unit may be configured to support a temperature gradient along the length of the flow channels, so that the temperature at the first end of the adsorbers is higher than the temperature at the second end of the adsorbers. The adsorbent in the adsorbers of the first PSA will include a first zone of adsorbent which is selective at the elevated operating temperature for carbon dioxide in preference to water vapour. Suitable such adsorbents known in the art include alumina impregnated with potassium carbonate, and hydrotalcite promoted with potassium carbonate.

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The adsorbent in the same or another zone of the adsorbers may include a component catalytically active at the operating temperature of that zone for the steam reforming reaction (e.g. fuel methane or methanol fuel) and for the water gas shift reaction. The catalytically active component in the second zone may be a reduced transition group metal or mixture of metals, or may be a transition group metal dispersed in zeolite cages and reversibly forming a metal carbonyl complex at the operating temperature of the second zone. Because carbon dioxide is preferentially adsorbed relative to steam, while enriched hydrogen is continually removed to the anode channel, the concentrations of carbon dioxide and hydrogen over the catalytically active component are maintained at a reduced level by the PSA process so as to shift the reaction equilibria favourably for the steam reforming and/or water gas shift reactions to proceed within the adsorbers of the first PSA unit. The conversion of carbon monoxide and reformable fuel components is driven toward completion to generate carbon dioxide and additional hydrogen. This is an example of a PSA reactor or Adsorption enhanced reactor®, enhancing the simple gas separation effect to further generate enriched hydrogen while removing the carbon dioxide, and driving the water gas shift reaction substantially to completion while achieving adequate purification of the hydrogen.

Industrial H₂ PSA is normally conducted at considerably elevated pressures (> 10 bars) to achieve simultaneous high purity and high recovery (~ 80% - 85%). Fuel cell systems operating with pressurized methanol reformers or in integration with gas turbine cycles may operate at relatively high pressures. Molten carbonate fuel cells operate at pressures up to about 10 bars.

At very low feed pressures (e.g. 2 - 3 bars), the first PSA would need supplemental compression to achieve high recovery of hydrogen and simultaneously high concentration of carbon dioxide. We may consider vacuum pumping to widen the working pressure ratio, or alternatively "heavy reflux" which is recompression and recycle to the PSA feed of a fraction of its exhaust stream at full pressure. Vacuum and heavy reflux options may be combined.

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The prior art includes combined cycle power plants with a gas turbine cycle integrated with a fuel cell system. Co-pending Canadian patent application No. 2,274,240 provides examples of how PSA units may be integrated with gas turbine power plants, or with fuel cell power plants having a gas turbine auxiliary engine.

5 The gas turbine may power all compressors and vacuum pumps for the O₂ PSA, along with vacuum pump and/or heavy reflux compression for the H₂ PSA. This auxiliary gas turbine cycle allows a heavy reflux vacuum pump and compressor to be driven by the turboexpander which expands the products of hydrogen PSA tail gas combustion. A feature here is integration of the vacuum pump(s) with the gas
10 turbine powered by tail gas combustion. Either single or multiple spool gas turbine configurations may be considered. Centrifugal or axial machines may be used as the compressors and pumps. Approaches based on integration of gas turbines and fuel cells are particularly favourable for larger power levels.

15 BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 shows an axial section of a rotary PSA module.

20 Figs. 2 through 5B show transverse sections of the module of Fig. 1.

Figs. 6 through 9 show simplified schematics of alternative molten carbonate fuel cell plants according to the invention.

25 DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

Figs. 1 - 5

Fig. 1 shows a rotary PSA module 1, which includes a number "N" of adsorbers 3 in adsorber housing body 4. Each adsorber has a first end 5 and a
30 second end 6, with a flow path therebetween contacting a nitrogen-selective adsorbent. The adsorbers are deployed in an axisymmetric array about axis 7 of the adsorber housing body. The housing body 4 is in relative rotary motion about

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axis 7 with first and second functional bodies 8 and 9, being engaged across a first valve face 10 with the first functional body 8 to which feed gas mixture is supplied and from which the heavy product is withdrawn, and across a second valve face 11 with the second functional body 9 from which the light product is withdrawn.

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In preferred embodiments as particularly depicted in Figs. 1 - 5, the adsorber housing 4 rotates and shall henceforth be referred to as the adsorber rotor 4, while the first and second functional bodies are stationary and together constitute a stator assembly 12 of the module. The first functional body shall henceforth be referred to as the first valve stator 8, and the second functional body shall henceforth be referred to as the second valve stator 9.

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In the embodiment shown in Figs. 1 - 5, the flow path through the adsorbers is parallel to axis 7, so that the flow direction is axial, while the first and second valve faces are shown as flat annular discs normal to axis 7. However, more generally the flow direction in the adsorbers may be axial or radial, and the first and second valve faces may be any figure of revolution centred on axis 7. The steps of the process and the functional compartments to be defined will be in the same angular relationship regardless of a radial or axial flow direction in the adsorbers.

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Figs. 2 - 5 are cross sections of module 1 in the planes defined by arrows 12 - 13, 14 - 15, and 16 - 17. Arrow 20 in each section shows the direction of rotation of the rotor 4.

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Fig. 2 shows section 12 B 13 across Fig.1, which crosses the adsorber rotor. Here, "N" = 72. The adsorbers 3 are mounted between outer wall 21 and inner wall 22 of adsorber wheel 208. Each adsorber comprises a rectangular flat pack 3 of adsorbent sheets 23, with spacers 24 between the sheets to define flow channels

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here in the axial direction. Separators 25 are provided between the adsorbers to fill void space and prevent leakage between the adsorbers.

5 As shown in Fig. 1, the adsorbers 3 comprise a plurality of distinct zones between the first end 5 and the second end 6 of the flow channels, here shown as three zones respectively a first zone 26 adjacent the first end 5, a second zone 27 in the middle of the adsorbers, and a third zone 28 adjacent the second end 6. The first zone typically contains an adsorbent or dessicant selected for removing very strongly adsorbed components of the feed gas mixture, such as water or methanol vapour, and some carbon dioxide. The second zone contains an adsorbent typically selected for bulk separation of impurities at relatively high concentration, and the third zone contains an adsorbent typically selected for polishing removal of impurities at relatively low concentration.

15 The adsorbent sheets comprise a reinforcement material, in preferred embodiments glass fibre, metal foil or wire mesh, to which the adsorbent material is attached with a suitable binder. For air separation to produce enriched oxygen, alumina gel may be used in the first zone to remove water vapour, while typical adsorbents in the second and third zones are X, A or chabazite type zeolites, typically exchanged with lithium, calcium, strontium, magnesium and/or other cations, and with optimized silicon/aluminum ratios as well known in the art. The zeolite crystals are bound with silica, clay and other binders, or self-bound, within the adsorbent sheet matrix.

25 Satisfactory adsorbent sheets have been made by coating a slurry of zeolite crystals with binder constituents onto the reinforcement material, with successful examples including nonwoven fibreglass scrims, woven metal fabrics, and expanded aluminum foils. Spacers are provided by printing or embossing the adsorbent sheet with a raised pattern, or by placing a fabricated spacer between adjacent pairs of adsorbent sheets. Alternative satisfactory spacers have been provided as woven metal screens, non-woven fibreglass scrims, and metal foils with etched flow channels in a photolithographic pattern.

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Typical experimental sheet thicknesses have been 150 microns, with spacer heights in the range of 100 to 150 microns, and adsorber flow channel length approximately 20 cm. Using X type zeolites, excellent performance has been achieved in oxygen separation from air at PSA cycle frequencies in the range of 30 to 150 cycles per minute.

Fig. 3 shows the porting of rotor 4 in the first and second valve faces respectively in the planes defined by arrows 14 - 15, and 16 - 17. An adsorber port 30 provides fluid communication directly from the first or second end of each adsorber to respectively the first or second valve face.

Fig. 4 shows the first stator valve face 100 of the first stator 8 in the first valve face 10, in the plane defined by arrows 14 - 15. Fluid connections are shown to a feed compressor 101 inducting feed air from inlet filter 102, and to an exhauster 103 delivering nitrogen-enriched second product to a second product delivery conduit 104. Compressor 101 and exhauster 103 are shown coupled to a drive motor 105.

Arrow 20 indicates the direction of rotation by the adsorber rotor. In the annular valve face between circumferential seals 106 and 107, the open area of first stator valve face 100 ported to the feed and exhaust compartments is indicated by clear angular segments 111 - 116 corresponding to the first functional ports communicating directly to functional compartments identified by the same reference numerals 111 - 116. The substantially closed area of valve face 100 between functional compartments is indicated by hatched sectors 118 and 119 which are slippers with zero clearance, or preferably a narrow clearance to reduce friction and wear without excessive leakage. Typical closed sector 118 provides a transition for an adsorber, between being open to compartment 114 and open to compartment 115. Gradual opening is provided by a tapering clearance channel between the slipper and the sealing face, so as to achieve gentle pressure equalization of an adsorber being opened to a new compartment. Much wider closed sectors (e.g. 119) are provided to substantially close flow to or from one end

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of the adsorbers when pressurization or blowdown is being performed from the other end.

5 The feed compressor provides feed gas to feed pressurization compartments 111 and 112, and to feed production compartment 113. Compartments 111 and 112 have successively increasing working pressures, while compartment 113 is at the higher working pressure of the PSA cycle. Compressor 101 may thus be a multistage or split stream compressor system delivering the appropriate volume of feed flow to each compartment so as to achieve the pressurization of adsorbers
10 through the intermediate pressure levels of compartments 111 and 112, and then the final pressurization and production through compartment 113. A split stream compressor system may be provided in series as a multistage compressor with interstage delivery ports; or as a plurality of compressors in parallel, each delivering feed gas to the working pressure of a compartment 111 to 113.
15 Alternatively, compressor 101 may deliver all the feed gas to the higher pressure, with throttling of some of that gas to supply feed pressurization compartments 111 and 112 at their respective intermediate pressures.

20 Similar, exhauster 103 exhausts heavy product gas from countercurrent blowdown compartments 114 and 115 at the successively decreasing working pressures of those compartments, and finally from exhaust compartment 116 which is at the lower pressure of the cycle. Similarly to compressor 101, exhauster 103 may be provided as a multistage or split stream machine, with stages in series or in parallel to accept each flow at the appropriate intermediate pressure descending to
25 the lower pressure.

30 In the example embodiment of Fig. 4A, the lower pressure is ambient pressure, so exhaust compartment 116 exhaust directly to heavy product delivery conduit 104. Exhauster 103 thus provides pressure letdown with energy recovery to assist motor 105 from the countercurrent blowdown compartments 114 and 115. For simplicity, exhauster 103 may be replaced by throttling orifices as

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countercurrent blowdown pressure letdown means from compartments 114 and 115.

5 In some preferred embodiments, the lower pressure of the PSA cycle is subatmospheric. Exhauster 103 is then provided as a vacuum pump, as shown in Fig. 4B. Again, the vacuum pump may be multistage or split stream, with
10 separate stages in series or in parallel, to accept countercurrent blowdown streams exiting their compartments at working pressures greater than the lower pressure which is the deepest vacuum pressure. In Fig. 4B, the early countercurrent blowdown stream from compartment 114 is released at ambient pressure directly
15 to heavy product delivery conduit 104. If for simplicity a single stage vacuum pump were used, the countercurrent blowdown stream from compartment 115 would be throttled down to the lower pressure over an orifice to join the stream from compartment 116 at the inlet of the vacuum pump.

15 Figs. 5A and 5B shows the second stator valve face, at section 16 - 17 of Fig. 1. Open ports of the valve face are second valve function ports communicating directly to a light product delivery compartment 121; a number of
20 light reflux exit compartments 122, 123, 124 and 125; and the same number of light reflux return compartments 126, 127, 128 and 129 within the second stator. The second valve function ports are in the annular ring defined by circumferential
25 seals 131 and 132. Each pair of light reflux exit and return compartments provides a stage of light reflux pressure letdown, respectively for the PSA process functions of supply to backfill, full or partial pressure equalization, and cocurrent blowdown to purge.

25 Illustrating the option of light reflux pressure letdown with energy recovery, a split stream light reflux expander 140 is shown in Figs. 1 and 5A to provide pressure let-down of four light reflux stages with energy recovery. The light reflux expander provides pressure let-down for each of four light reflux stages, respectively between
30 light reflux exit and return compartments 122 and 129, 123 and 128, 124 and 127, and 125 and 126 as illustrated. The light reflux expander 140 may power a light product booster compressor 145 by drive shaft 246, which delivers the oxygen enriched light

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product to oxygen delivery conduit 147 and compressed to a delivery pressure above the higher pressure of the PSA cycle. Illustrating the option of light reflux pressure letdown with energy recovery, a split stream light reflux expander 240 is provided to provide pressure let-down of four light reflux stages with energy recovery. The light
5 reflux expander serves as pressure let-down means for each of four light reflux stages, respectively between light reflux exit and return compartments 122 and 129, 123 and 128, 124 and 127, and 125 and 126 as illustrated.

Light reflux expander 240 may be coupled to a pressure booster compressor
10 245 by drive shaft 246. Compressor 245 receives the light product from conduit 25, and delivers light product (compressed to a delivery pressure above the higher pressure of the PSA cycle) to delivery conduit 250. Alternatively, compressor 245 could be applied to recirculation of anode gas as shown in Fig. 7. Since the light reflux and light product have approximately the same purity, expander 140 and light product
15 compressor 145 may be hermetically enclosed in a single housing which may conveniently be integrated with the second stator as shown in Fig. 1. This configuration of a "turbocompressor" booster without a separate drive motor is advantageous, as a useful pressure boost can be achieved without an external motor and corresponding shaft seals, and can also be very compact when designed to operate
20 at high shaft speeds.

Fig. 5B shows the simpler alternative of using a throttle orifice 150 as the pressure letdown means for each of the light reflux stages.

25 Turning back to Fig. 1, compressed feed gas is supplied to compartment 113 as indicated by arrow 125, while heavy product is exhausted from compartment 117 as indicated by arrow 126. The rotor is supported by bearing 160 with shaft seal 161 on rotor drive shaft 162 in the first stator 8, which is integrally assembled with the first and second valve stators. The adsorber rotor is driven by motor 163 as rotor drive
30 means.

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A buffer seal 170 is provided to provide more positive sealing of a buffer chamber 171 between seals 131 and 171. In order to further minimize leakage and to reduce seal frictional torque, buffer seal 171 seals on a sealing face 172 at a much smaller diameter than the diameter of circumferential seal 131. Buffer seal 170 seals between a rotor extension 175 of adsorber rotor 4 and the sealing face 172 on the second valve stator 9, with rotor extension 175 enveloping the rear portion of second valve stator 9 to form buffer chamber 171. A stator housing member 180 is provided as structural connection between first valve stator 8 and second valve stator 9.

In the following figures of this disclosure, simplified diagrams will represent the PSA apparatus as described above. These highly simplified diagrams will indicate just a single feed conduit 181 to, and a single heavy product conduit 182 from, the first valve face 10; and the light product delivery conduit 147 and a single representative light reflux stage 184 with pressure let-down means communicating to the second valve face 11.

Figs. 6 - 9

Each of Figs. 6 - 9 is a simplified schematic of a molten carbonate fuel cell power plant 200, including the fuel cell 202, a high temperature PSA unit 204 cooperating with a combustor 206 to transfer carbon dioxide from the anode side to the cathode side of the fuel cell, and an integrated gas turbine unit 208 for gas compression and expansion. The PSA unit 204 increases hydrogen concentration and reduces carbon dioxide concentration over the cathode, thus increasing cell voltage. This directly increases fuel cell stack efficiency and electrical output, while also reducing the heat generated by the fuel cell so that the fraction of plant power output to be recovered less efficiently by a thermal bottoming cycle is reduced.

Fuel cell 202 includes the molten carbonate electrolyte 210 supported on a porous ceramic matrix, interposed between anode channel 212 and cathode channel

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214. The anode channel has an inlet 216 and an outlet 218, while the cathode channel 214 has an inlet 220 and an outlet 222.

The embodiment of Fig. 6 illustrates two alternatives for feed gas supply in combination. More typically, either of these feed gas supply alternatives might be used separately in any given MCFC installation. These alternatives correspond to the suitability of the feed gas for direct admission to the fuel cell anode, or for admission only after treatment by the first PSA unit. For the case of natural gas being the fuel, these alternatives also correspond to the fuel processing options or combinations of (1) "internal reforming" within the fuel cell stack, (2) "sorption enhanced reforming" within the first PSA unit, or (3) "external reforming" outside the immediate MCFC system as here described.

Endothermic reforming reactions are $\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2$
 and $\text{CH}_4 + 2\text{H}_2\text{O} \rightarrow \text{CO}_2 + 4\text{H}_2$,
 with exothermic water gas shift $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$,
 supplemented by partial combustion in the case of autothermal reforming
 $\text{CH}_4 + 2\text{O}_2 \rightarrow \text{CO} + 2\text{H}_2$.

A first feed gas supply inlet 230 communicates to anode inlet 214, introducing a first feed gas already compressed and preheated to the MCFC working conditions. The first feed gas might be hydrogen, syngas generated by an external fuel processor (e.g. coal gasifier or steam methane reformer), or natural gas for internal reforming within the anode channel 212 which would then be modified as known in the art to contain a suitable steam reforming catalysts such as nickel supported on alumina.

A second feed gas supply inlet 240 communicates to a feed production compartment in first rotary valve face 10 of the first PSA unit 204, again introducing feed gas already compressed and preheated to the first PSA unit higher pressure and working temperature. The carbon dioxide and steam enriched heavy product stream is released from the blowdown and exhaust compartments in first

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rotary valve face 10 into conduit 242 at the lower pressure of the first unit PSA cycle. The higher pressure of the first PSA is slightly above the working pressure of the MCFC, while the lower pressure may be atmospheric or even subatmospheric.

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The heavy product stream from conduit 242 is compressed back up to the higher pressure of first PSA by carbon dioxide compressor 244, which delivers the compressed heavy product stream to conduit 246 which branches to heavy reflux conduit 247 communicating to a feed production compartment in first rotary valve face 10 of the first PSA unit 204, and to gas turbine combustor 206.

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Enriched hydrogen light product gas from first PSA 204 is delivered by conduit 250 from the second rotary valve face 11 of the first PSA unit to anode inlet 216. Three stages of light reflux are shown, in which separate streams of light product gas at successively declining pressures are withdrawn from the second rotary valve face 11 for pressure letdown in respective stages of light reflux expander 140, and then returned to the second rotary valve face for purging and repressurization of the adsorbers. After passing through the anode channel 212, anode gas depleted in hydrogen and enriched in carbon dioxide and steam is withdrawn from anode exit 218 through conduit 255 or treatment by first PSA unit 204 to recover hydrogen while removing carbon dioxide and at least a portion of the steam.

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Anode channel 212, conduit 255, the PSA unit 204, and conduit 250 comprise an anode loop in which hydrogen is recirculated and replenished for substantially complete utilization, while carbon dioxide is continually removed by PSA 204. A pressure booster means is required to overcome flow pressure drop around the anode loop. In Fig. 6, the pressure booster means is the PSA unit 204, with no mechanical pressure booster being required. The anode exhaust gas in conduit 255 is at a moderately lower pressure than the feed gas in infeed conduit 240 and the heavy reflux gas in conduit 247. Hence the anode exhaust gas is introduced to a feed repressurization compartment in first rotary valve face 10.

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After the anode exhaust gas has entered the adsorbers 3, it is there compressed back up to the higher pressure by feed gas and heavy reflux gas entering the adsorbers from conduits 240 and 247.

5 Gas turbine assembly 208 includes compressor 260 and turbine 262, coupled to a motor/generator 264 by shaft 266, and to heavy product compressor 244 and light reflux expander 140 by shaft 267. Ambient air is introduced to compressor 260 by infeed conduit 270, and is there compressed to working pressure for delivery by conduit 272 to combustor 206. Combustor 206 burns residual fuel
10 values (including some hydrogen and unconverted carbon monoxide and fuel) in the carbon dioxide rich heavy product stream. A catalyst may be provided in combustor 206 to ensure stable combustion with high inert concentrations, or supplemental fuel may be added thereto. The hot gas exiting combustor 206 by conduit 280 is cooled in recuperative heat exchanger 285 to approximately the
15 MCFC operating temperature for admission as cathode gas to cathode inlet 220. The cathode gas contains carbon dioxide and residual oxygen, diluted by steam and nitrogen. After circulation through cathode channel 214 in which some oxygen and carbon dioxide are consumed, the depleted cathode gas is conveyed from cathode exit 222 by conduit 290 back to recuperator 285 for reheat to an elevated turbine
20 entry temperature for admission by conduit 291 to turbine 262. After expansion through turbine 262, the exhaust cathode gas is discharged through conduit 292 where further heat exchange would preferably take place to obtain most efficient heat recovery, e.g. for preheating the feed gas to inlets 230 and 240.

25 Several alternative features and improvements are shown in Fig. 7. In this figure, a thermally integrated reformer is shown. Already compressed fuel and water (or steam) are admitted from infeed conduit 300, passing through an exhaust recuperator 302 for recovering heat from expanded cathode exhaust in conduit 292, and then passing through recuperator 285 to reach an elevated reforming
30 temperature (e.g. 800° to 1200° C) for admission to catalytic reforming reactor 310. The endothermic reforming reaction reduces the temperature of the delivered

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syngas to about the MCFC temperature, and this syngas is delivered by conduit 240 to a feed production compartment in the first rotary valve face 10 of PSA unit 204.

5 An improvement not shown in Fig. 6 is the removal of water from the heavy product in conduit 242, prior to compression by carbon dioxide compressor 244. A condenser 320 is provided in conduit 242 for water removal and for cooling the heavy product gas so as to reduce the compression power required by compressor 244. Liquid water is removed by drain 321. The condensation temperature is established by cooler 322. A heat exchanger 324 is provided for
10 recuperative heat exchange between conduits 242 and 246.

A further feature in Fig. 7 is the provision of a mechanical pressure booster for the anode loop, as booster compressor 330 which is powered direction by light reflux expander 140 through shaft 267. Recompressed anode gas from conduit 255
15 is boosted back to the higher pressure by booster 330, and is delivered by conduit 331 to a production feed compartment in the first rotary valve face 10. A portion of the anode exhaust gas in conduit 255 may still be delivered directly to a feed pressurization compartment by conduit 333. In this example, the sole power source for booster 330 is expander 140, which is now separated from gas turbine
20 assembly 308.

In Figs. 8 and 9, further embodiments are shown incorporating an oxygen VPSA in order to boost the oxygen and carbon dioxide partial pressures in the cathode channel, so as to increase the cell electromotive force and thus reduce the
25 thermal bottoming load while enhancing overall plant efficiency. In Figs. 8 and 9 as in Fig. 6, various details of recuperative heat recovery and water condensation from the heavy product that were shown in Fig. 7 are omitted for simplicity and clarity, though it will be understood that these features are preferably included.

30 The oxygen PSA or VPSA unit 400 include a rotary module 401 with nitrogen-selective adsorbent in adsorbers 403, a first rotary valve face 410 and a second rotary valve face 411. The first rotary valve face 410 receives compressed

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feed air at a feed production compartment from feed air compressor 260 via conduit 420, and discharges exhaust nitrogen enriched air from an exhaust compartment via conduit 422 to an optional vacuum pump 424 (to be included for VPSA or excluded for simple PSA) for discharge to atmosphere or any other use for moderately enriched nitrogen. The second rotary valve face 411 delivers enriched light product oxygen at e.g. 90% purity by non-return valve 430 in conduit 431 delivering the oxygen to oxygen compressor 432 which delivers the oxygen at least the MCFC working pressure to conduit 434 and thence combustor 206. Light reflux pressure letdown throttles 436 are also provided for light reflux stages in the second rotary valve face 411.

Oxygen enrichment of the air provided to combustor 206 reduces the inert load of nitrogen in the cathode channel, thus enhancing electrochemical energy conversion performance as discussed above. The working fluid for gas turbine expander is thus largely concentrated carbon dioxide with only small amounts of atmospheric gases.

Fig. 9 shows the additional feature that a portion of the enriched oxygen from PSA 400 is used for fuel processing, either within the plant as here shown, or externally as in the example that coal gasification is used to generate syngas feed. Here, a portion of the compressed oxygen in conduit 434 is conveyed by conduit 440 to reformer 310, which here is an autothermal reformer for e.g. steam reforming natural gas.

Fig. 2

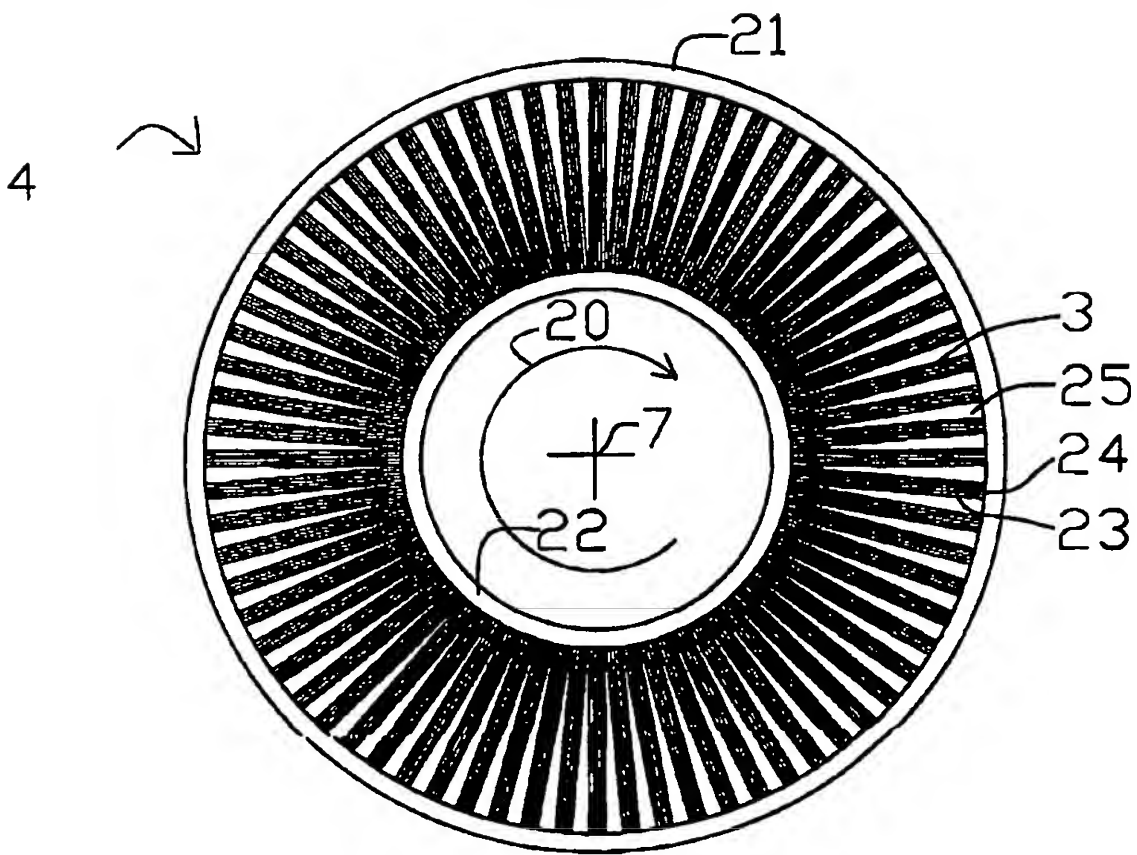


Fig. 3

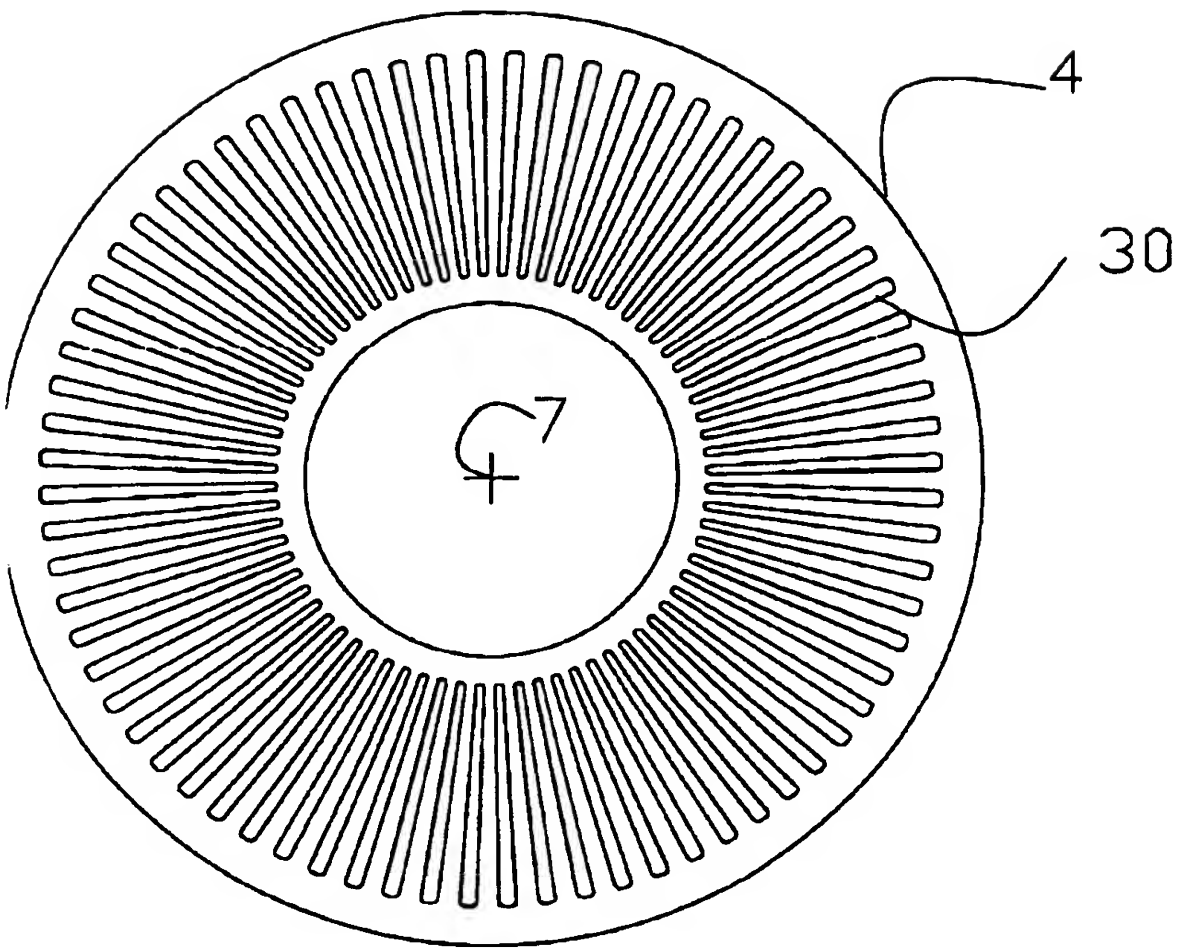


Fig. 4A

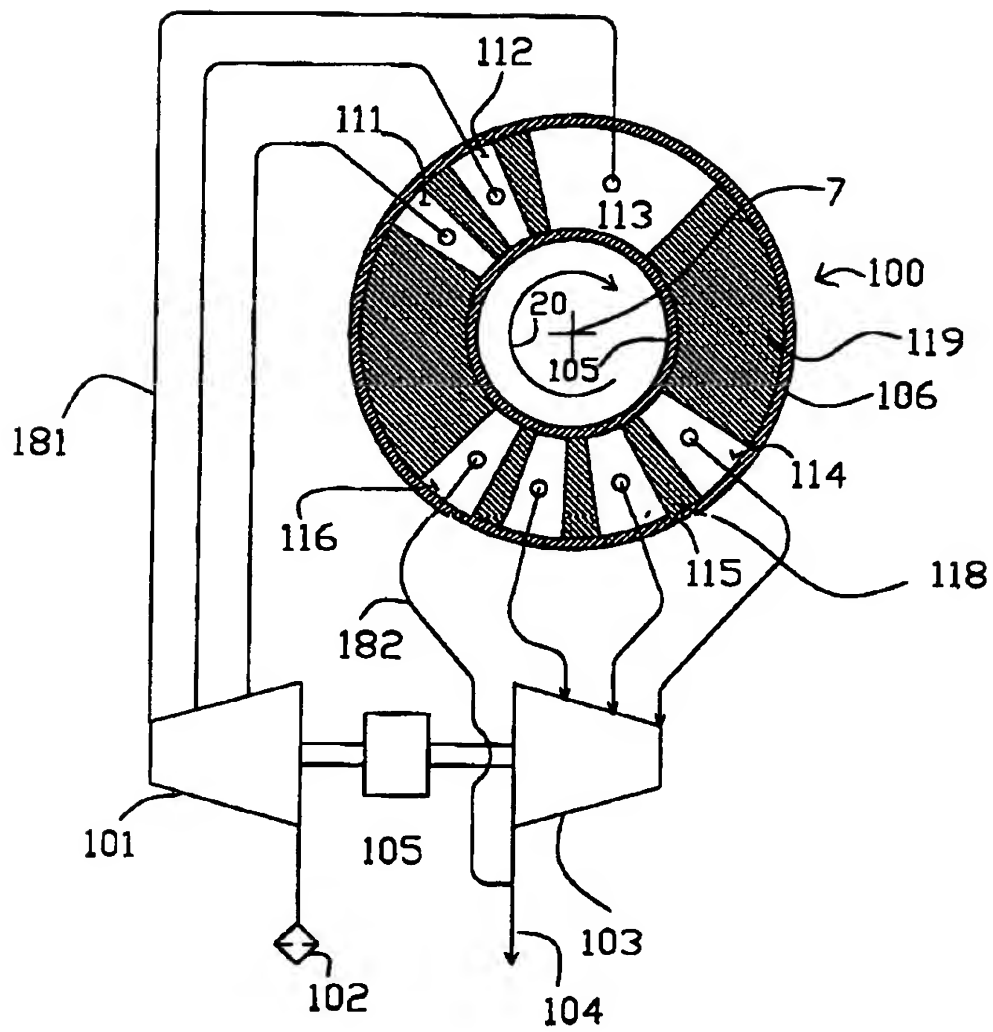


Fig. 4B

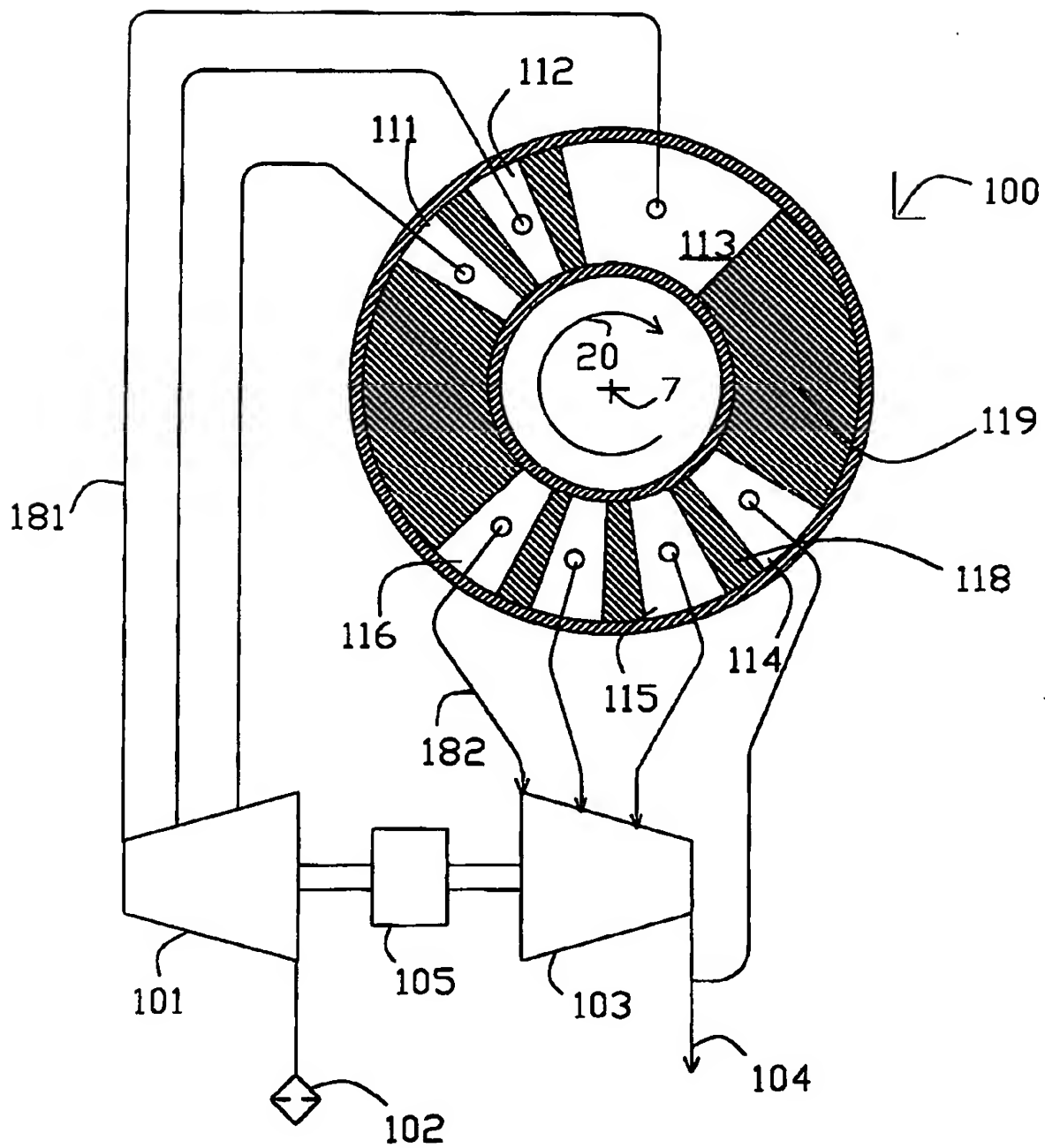


Fig. 5A

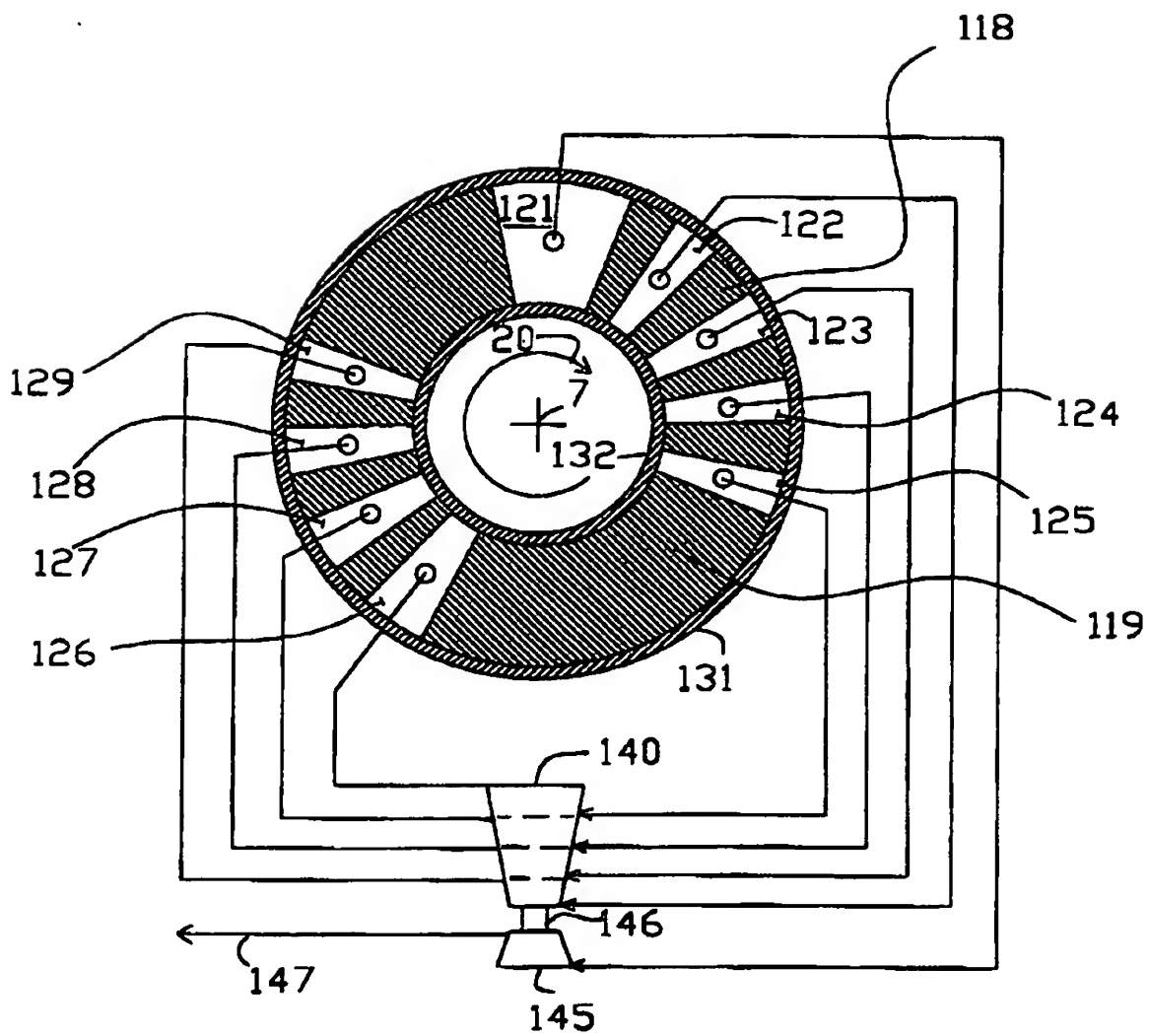


Fig. 5B

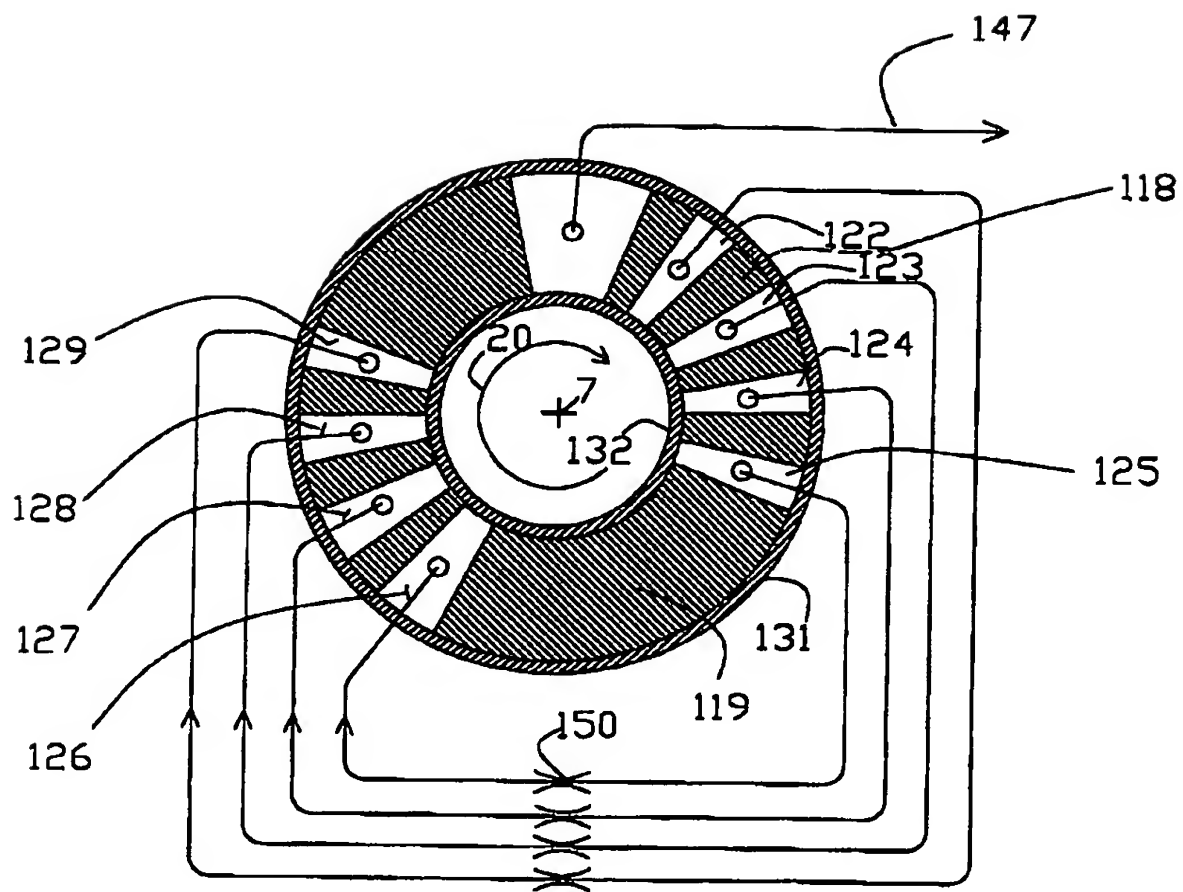


FIG. 6

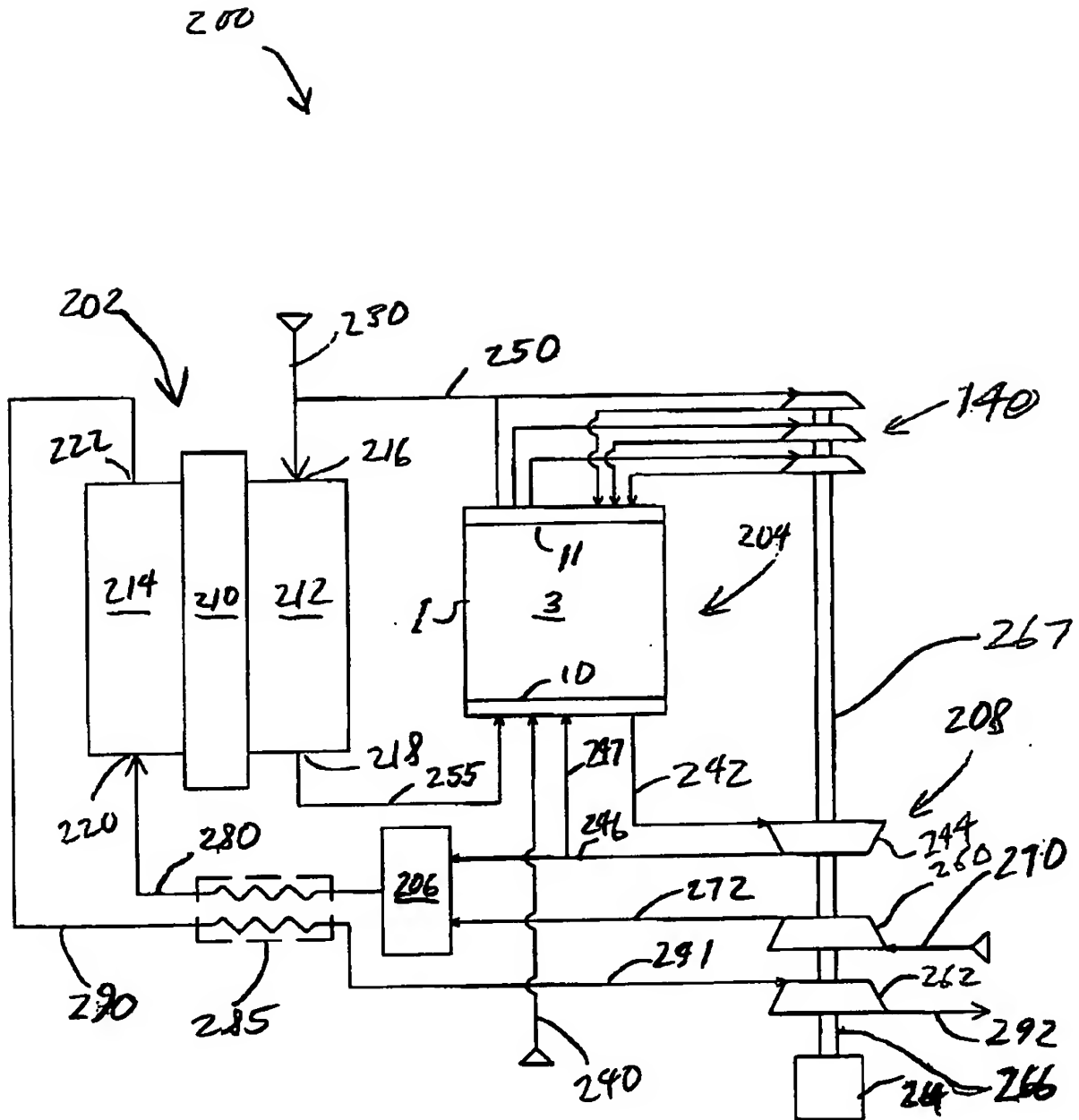


FIG. 7

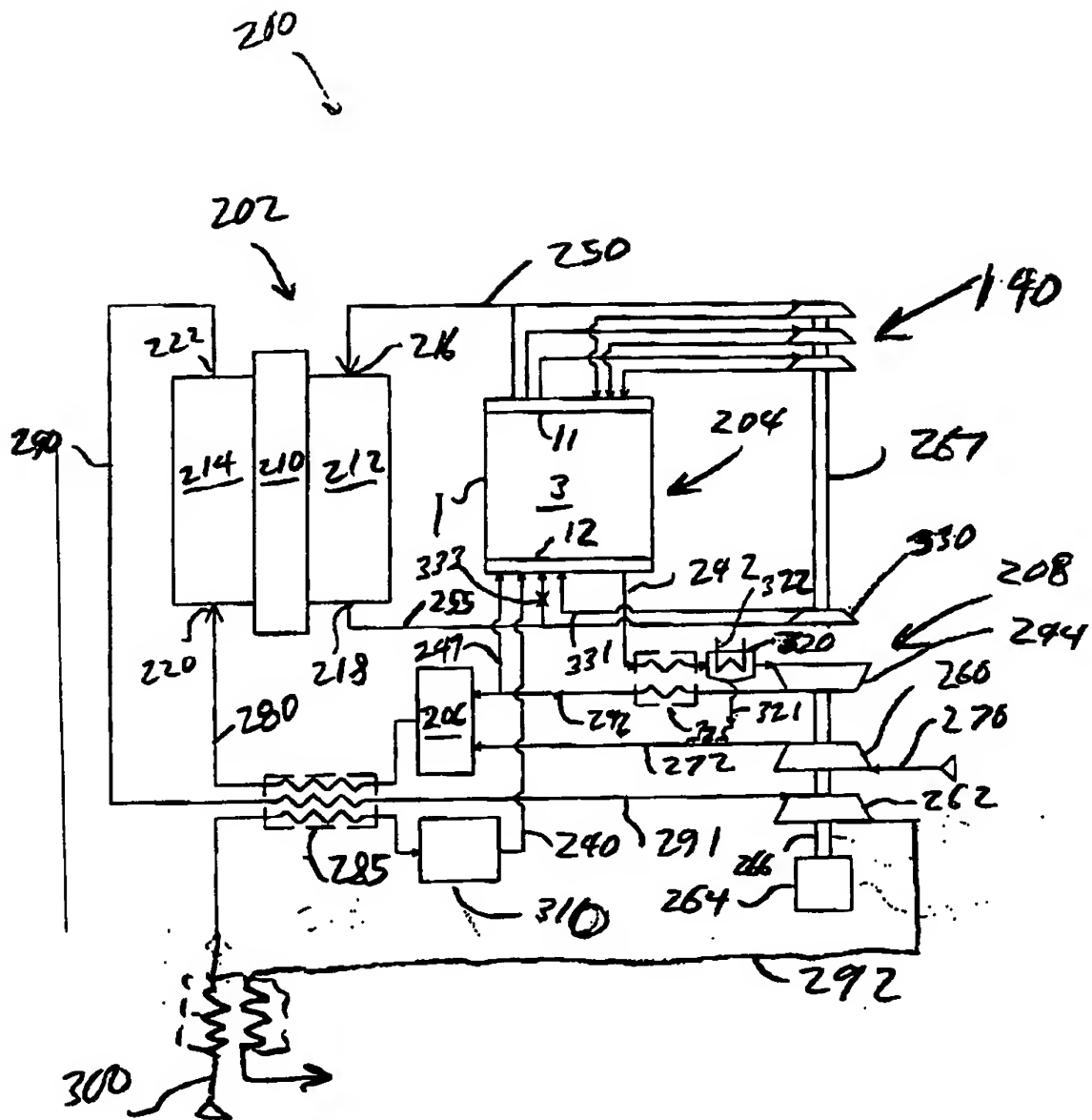


FIG. 8

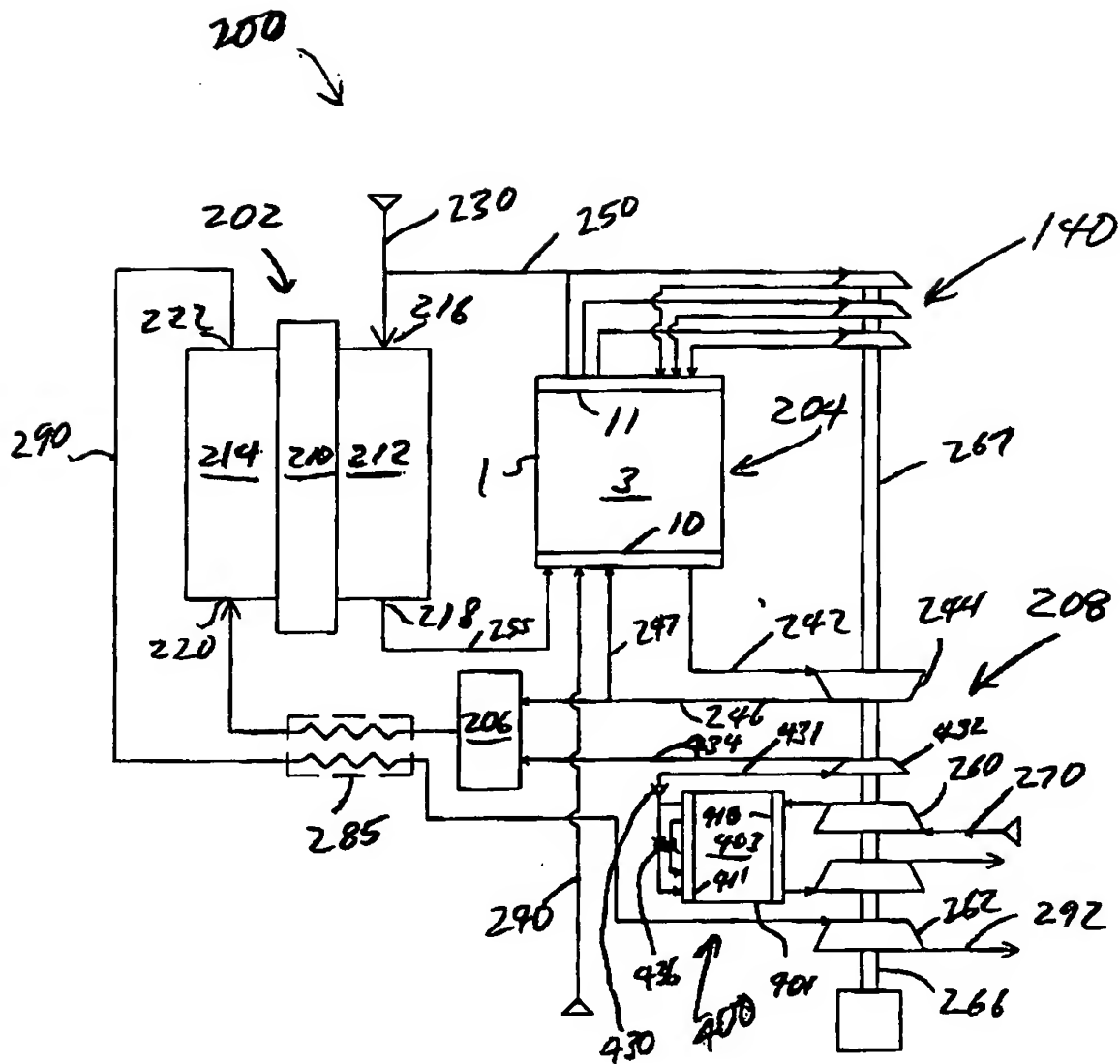


FIG. 9

